

Dark-state mechanism for the long-time transfer of excitations in a donor–acceptor system

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ABSTRACT

The excitation energy transfer between a donor–acceptor pair with fixed distance apart through energy exchanging with environment is investigated. The total system is modeled as two two-level systems (TLSs) interacting with many harmonic oscillators. The pair behaves coherently or incoherently, depending on whether the dipolar coupling is stronger or weaker than the TLS–environment coupling. The environmental linear dispersion relation gives an analytical solution to the pair's probability involving all the retardation times. We found that the long-time trapping of energy within the pair is caused by the inhibiting dark-state radiative decay when two TLSs are at half a resonant wavelength.

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1. Introduction

Energy sources are the lifeblood of modern society. To avoid depleting our natural resources and destroying the environment, there is keen to seek for clean energy, particularly, solar power – the available renewable energy on earth. Studies on the excitation energy transfer in plants, algae and bacteria [1] over the past years have shed light on creating a fully integrated energy system. For a long time in the past, it has been believed that biological process is classical. However, recent experimental evidences have shown that light-harvesting complexes may harness quantum-mechanical effects such as the surprisingly long-lasting coherence in photosynthesis [2–4]. Therefore, excitation energy transfer assisted by quantum coherence attracts considerable attention recently [5–13]. So far the mechanism of the light-harvesting process is still elusive, and scientists continuously try to understand how light harvesting complexes can capture and transport light energy with near-unit efficiency from theoretical aspects, such as the symmetry of the initial quantum states [8,9], the structure and arrangement of antenna complexes [10], the accumulative quantum phases in closed transfer pathways [11,12], the emergence of correlated energetic fluctuations between different chromophores [14–19]. Ongoing researches inspire considerable efforts to create artificial systems for quantum transport and optimized solar cells in semiconductor fabrication [20–23].

The donor–acceptor model serves as a toy model to study the dynamics of the excitation energy transfer in the presence of environments. To understand the correlation between fluctuations in the donor–acceptor pair and explore the potential applications in modern engineering technique and manufacture, we consider the quantum coherence transfer of excitation energy between a donor and an acceptor with fixed distance d apart. In contrary to the popular assumption of independent environments for donor and acceptor, we assume that the donor and acceptor share an environment. It is well known that the environment causes two distinct effects on the quantum system: quantum dissipation and decoherence. Only quantum dissipation allows energy exchange between the system and environment. So we consider the effect of quantum dissipation on the quantum coherence transfer of excitations between a donor–acceptor pair. In our model, the donor and acceptor are modeled as two two-level systems (TLSs) and the environment is modeled as many harmonic oscillators [24]. The general treatment of this problem for an arbitrary line shape of the reservoir coupling spectrum is achieved by the Laplace transformation under Wigner–Weisskopf approximation in one quantum subspace. An analytical solution to the probability amplitudes of the pair is obtained by taking the linear expansion of the environmental frequency. In our approach, it is not necessary to assume whether the dipole coupling is weaker or stronger than the pair–environment coupling.

This Letter is organized as follows. In Section 2, we present our model. In Section 3, a general approach is given in the one quantum subspace where the linewidths of the donor and acceptor are implied in the self-energies and the position-dependent self-energies. In Section 4, we evaluate the probability amplitude

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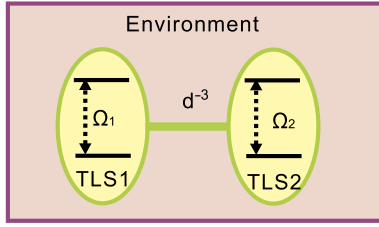


Fig. 1. (Color online.) Sketch of a donor–acceptor pair embedded in an environment. The donor and acceptor are modeled as two two-level systems, which interact with each other by the dipole–dipole interaction, while the environment is treated as many oscillators with frequencies ω_k .

of finding either the donor or the acceptor on its excited state by taking the linear expansion of the environmental frequency. We draw our conclusions in Section 5.

2. Description of the model

Consider a donor–acceptor pair embedded in a common environment. Both donor and acceptor are modeled as two two-level systems with fixed distance d apart, as shown in Fig. 1, where TLS1 and TLS2 represent the donor and acceptor respectively. Each TLS has a ground state $|g_i\rangle$, an excited state $|e_i\rangle$ and energy spacing Ω_i . Define σ_i^+ and σ_i^- as the raising and lowering pseudo-spin operators for the i th TLS. The Hamiltonian H_S of the donor–acceptor pair is the sum of the individual TLS Hamiltonian and dipole–dipole interaction Hamiltonian

$$H_S = \sum_{i=1,2} \hbar \frac{\Omega_i}{2} \sigma_i^z + \hbar J (\sigma_1^+ \sigma_2^- + \sigma_2^+ \sigma_1^-), \tag{1}$$

where $J = V/d^3$ is the dipole–dipole coupling strength.

The environment consists of many oscillators with frequencies ω_k , whose Hamiltonian reads

$$H_E = \sum_k \hbar \omega_k a_k^\dagger a_k, \tag{2}$$

where the creation and annihilation operators a_k^\dagger and a_k satisfy the bosonic commutation relation $[a_k, a_{k'}^\dagger] = \delta_{kk'}$. In most natural condition, only single excitation occurs [25]. Therefore, one can regard the environment as a field described by the plane-wave modes associated with operators a_k^\dagger and a_k . The TLS–environment interaction is of the “electric-dipole form”. In the rotating-wave approximation, the TLS–environment Hamiltonian reads

$$H_{SE} = \sum_{ki} \hbar (g_{ik} \sigma_i^+ a_k + g_{ik}^* \sigma_i^- a_k^\dagger), \tag{3}$$

where the coupling strength $g_{ik} = |g_{ik}| e^{ikx_i}$ is dependent on the location of the i th TLS. Here, we assume the donor and acceptor are located at position x_1 and x_2 , respectively. Their inter-distance $d \equiv |x_1 - x_2|$. The term e^{ikx_i} in the coupling strength g_{ik} leads to correlation between the donor and acceptor via the environment. The total system is described by the Hamiltonian

$$H = H_S + H_E + H_{SE}. \tag{4}$$

The system described by Eq. (4) could be realized by two impure atoms inside a crystal. The lattice vibrational modes serve as the environment. The dipole–dipole interaction comes from the Coulomb interaction between the electrons of the donor and acceptor which is expanded in a multiple series up to powers d^{-3} .

3. Formulation for single excitation

The single-excitation subspace is spanned by the states $\{|eg_0\rangle, |ge_0\rangle, |gg1_k\rangle\}$, where $\{|gg1_k\rangle\}$ represents the state with one excitation in the k th mode of the environment and both TLSs on the ground state, while $|D\rangle \equiv |eg_0\rangle$ and $|A\rangle \equiv |ge_0\rangle$ represent states with a single excitation in one of the TLSs. The state of the system at time t can be expressed as

$$|\psi(t)\rangle = e^{-i\Omega t} (C_1 |D\rangle + C_2 |A\rangle) + \sum_k A_k e^{-i\omega_k t} |gg1_k\rangle, \tag{5}$$

where $\Omega \equiv (\Omega_1 + \Omega_2)/2$ is the average energy spacing of the pair. C_i and A_k are the time-dependent probability amplitudes for finding the system on the corresponding state.

Inserting the state (5) into Schrödinger equation, we obtain the evolution equations for the amplitudes as

$$\partial_t C_1 = -i \frac{\Delta}{2} C_1 - i J C_2 - i \sum_k g_{1k} A_k e^{-i(\omega_k - \Omega)t}, \tag{6a}$$

$$\partial_t C_2 = i \frac{\Delta}{2} C_2 - i J C_1 - i \sum_k g_{2k} A_k e^{-i(\omega_k - \Omega)t}, \tag{6b}$$

$$\partial_t A_k = -i (g_{1k}^* C_1 + g_{2k}^* C_2) e^{i(\omega_k - \Omega)t}, \tag{6c}$$

where $\Delta = \Omega_1 - \Omega_2$ is the energy detuning between TLS1 and TLS2. Integrating Eq. (6c) for A_k , and then inserting it into Eq. (6a), (6b) for C_i , we obtain the exact integro-differential equations for C_i . We then perform Laplace transform to the integro-differential equations. With the initial conditions $C_i(t=0) \equiv C_{i0}$, we obtain

$$s \tilde{C}_1 = C_{10} - i \frac{\Delta}{2} \tilde{C}_1 - \tilde{C}_1 \Phi_1(s) - [iJ + \Psi(s)] \tilde{C}_2, \tag{7a}$$

$$s \tilde{C}_2 = C_{20} + i \frac{\Delta}{2} \tilde{C}_2 - \tilde{C}_2 \Phi_2(s) - [iJ + \Psi(s)] \tilde{C}_1, \tag{7b}$$

where \tilde{C}_i denotes the Laplace transform of $C_i(t)$. In the above, we have introduced

$$\Phi_i(s) = \sum_k \frac{|g_{ik}|^2}{s + i(\omega_k - \Omega)}, \tag{8}$$

$$\Psi(s) = \sum_k \frac{g_{2k}^* g_{1k}}{s + i(\omega_k - \Omega)}. \tag{9}$$

The function $\Phi_i(s)$ describes the self-energy of the excited state of the i th TLS and the ground state of the other TLS. The function $\Psi(s)$ describes the x -dependent self-energy. Actually, the function $\Phi_i(s)$ is related to the reservoir coupling spectrum

$$\Gamma_i(\omega) = \sum_k \pi |g_{ik}|^2 \delta(\omega - \omega_k), \tag{10}$$

which characterizes the spectral shape of the reservoir. Since the states in the environment are very dense (continuum), one can replace the summation by integral. In the spirit of the Weisskopf–Wigner approximation [26] or pole approximation [27], where the environment is assumed to be spectrally flat and the frequencies of the TLS is assumed to be deeply embedded in the continuum, we neglect the z -dependence of function $\Phi_i(\epsilon + iz)$ by letting $z=0$ [27]

$$\Gamma_i(\Omega) = \Phi_i(\epsilon + iz)|_{z=0} = \left(\frac{\eta \pi |g_{ik}|^2}{\partial_k \omega_k} \right)_{k=k_0}, \tag{11}$$

where the wave number k_0 is the solution of $\omega_k = \Omega$ and the constant η is a dimension-dependent parameter (e.g. for one dimension, $\eta = L/(2\pi)$). Hereafter, for convenience, we will let

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